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(54) **THERMOPLASTIC COMPOSITE MATERIAL REINFORCED WITH HEMP FIBERS.**

(57) A novel thermoplastic composite material which is reinforced with 'hemp fibers' and which shows high impact strength, high tensile strength, high bending modulus and high heat deformation temperature. This composite material is obtained by mixing fibers of hemp such as ramie, flax, hemp, jute, Manila hemp or sisal hemp with a wooden material such as wood flour or wood particles to prepare a homogeneous mixture, adding thereto a thermoplastic matrix such as polypropylene or polyethylene, well mixing the total components to homogeneously disperse the hemp fibers and the wooden material in the matrix and form composite particles. The composite particles are then formed into sheets adapted for stamping or pellets adapted for injection molding or transfer molding.

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COMPOSITE THERMOPLASTIC MATERIALS REINFORCED WITH HEMP FIBERS

Technical Field

This invention relates to novel composite thermoplastic materials reinforced with "hemp fibers" so as to have a high degree of impact strength, thermostability and moldability and more particularly to such composite thermoplastic materials suited particularly to use for manufacturing automobile interior fittings of light weight and low cost and the like.

Background Art

It is known that some composite thermoplastic materials such as those prepared by mixing thermoplastic matrix such as polypropylene and wood chips or wood dusts and forming the resultant mixture into the form of sheets or pellets are cheaper and lighter in weight as compared to plastic itself and advantageously employed for stamping or injection molding techniques to manufacture various articles including interior fittings of automobiles such as instrument panels, heater ducts, and so on.

An example of composite thermoplastic materials comprising polyolefins and wood chips is disclosed in Japanese Patent Publication SHO 60-40965 "Method and Apparatus for manufacturing panels" [field on March 22, 1974 claiming Convention Priority based on Swiss application No. 4249/73, by GOR, Italy, published on

September 13, 1985 (SHO 60)] in which about 40 to 60% by weight of polyolefins such as polypropylene and polyethylene and about 60 to 40% by weight of particulate cellulosic fillers are mixed together to form a composite thermoplastic material.

It is found, however, that particulate cellulosic fillers as taught in that publication are virtually difficult to be kept uniform as to their sizes and shapes and are not able to be uniformly dispersed in such materials as thermoplastic matrices which have rather high melt viscosity and the resulting composites cannot have sufficient uniformity in quality and physical properties throughout their entire mass. Such non-uniform composites may lead to production of rejects through stamping or injection molding processes due to varying and uneven physical properties and moldability. These composites having no assurance of uniformity inadvantageously require to adopt wide safety standards in quality control and product designing.

Though particulate cellulosic fillers are able to be more easily handled than filamentary reinforcing materials, they are inferior to that in impact strength, thermal distortion temperature, deep-drawing ability, and so on. Thus, thermoplastic composites reinforced with particulate cellulosic fillers have a limited degree of freedom in designing for production of thin wall articles, and have limited usages.

In an attempt to overcome these defects, it is proposed to use particulate cellulosic fillers in combination with a fibrous

reinforcing material. Japanese Laid-open publication SHO 59-207966 "Composite materials utilizing synthetic fiber rag" [filed on May 13, 1983 by K.K. Mikuni Mfg., Japan, provisionally published on November 26, 1984 (SHO 59)] shows this proposal in which Claim 2 defines a composite material comprising about 70 to 160 parts by weight of thermoplastic synthetic resins such as polypropylene, polyethylene and ABS, 100 parts by weight of rags of synthetic fiber such as polyester filament, and 50 parts by weight of wood chips or wastepaper. However, since synthetic fibers like polyester, being thermoplastic in nature, has low rigidity and low resistance to thermal deformation, it seems that there is no significant reinforcing effect attained by synthetic fibers except that is may contribute to lowering costs of composites. In other words, synthetic fiber rags can scarcely compensate for defects of particulate ligneous fillers and it will worsen physical properties of composites in greatly reducing bending resistance thereof upon heated. Thus, sheetings made from composite material containing synthetic fiber will be softened upon heated during stamping process such that it cannot be supported by operator's hands in a flattened state. Further, synthetic fibers will lower specific heat capacity of the composites to cause heat radiation therefrom to be rapid. For this reason, it will be difficult to handle composite sheetings in double layers. Besides, accurate control over molding conditions and/or precise process control will cumbersomely be required in using composite materials containing synthetic fiber reinforcement.

Disclosure of the Invention

It is an object of the present invention to provide novel composite thermoplastic materials which are of high quality and uniform in physical properties and does not undergo losses of strength, rigidity and specific heat capacity upon heated.

It is another object of the invention to provided composite thermoplastic materials which are light in weight and have high degrees of impact strength, thermostability and moldability.

It is yet another object of the invention to provide composite thermoplastic materials having characteristics as enumerated above and worked in the form of sheets for use in stamping process or pellets for use in injection or transfer molding processes.

It is a specific object of the present invention to provide composite thermoplastic sheetings capable of being easily handled in stamping molding processes and formed into complicated configurations including deep-drawn shapes.

These and other objects of the invention are attained by providing novel composite thermoplastic materials comprising:

(1) thermoplastic metrices such as polypropylene, polyethylene, nylon, acrylonitrile-butadiene-styrene copolymer (ABS) and so on,

(2) "hemp fibers" in the form of single fibers which are rigid, stiff, thermostable and yet lightweight, and

(3) ligneous fillers in the form of particles, dusts or scales, the latter two constituents being uniformly dispersed in the plastic matrices.

According to the invention, it is noted that single fibers selected from the group consisting of ramie (Boehmeria nivea), flax (Linum usitatissimum), hemp (Cannabis sativa), jute (Corchorus capsularis), manila hemp (Musa textilis), and sisal hemp (Agave sisalana) can advantageously be employed as novel reinforcing materials for plastics, and these fibers are generally referred to as "hemp fibers" in this specification and claims for convenience of explanation. Latin in each parenthesis indicates a botanical name of the plant from which each fiber preceding the parenthesis is obtained.

"Hemp fibers" are noted to have substantial uniformity in shapes and be able to relatively easily be made all of uniform length. Moreover, they have a good compatibility with particulate ligneous or cellulosic fillers employed heretofore, especially when adequate mixing or kneading processes are utilized. Thus, hemp fibers accompanying ligneous fillers distributed uniformly therein can be added to the thermoplastic matrices in a manner that both hemp fibers and ligneous fillers are substantially uniformly dispersed int he matrices, which otherwise would not be attained by adding ligneous fillers alone.

Hemp fibers employed in this invention have good characteristics such as light weight, high rigidity and high thermal resistance, and can be optimum reinforcing fibers to impart to the composites high degrees of impact and tensile strengths which ~~e~~annot be attained by using particulate ligneous cellulosic

fillers alone. Hemp fibers can also improve largely thermal distortion temperatures and deep-drawing capability of the resultant composites and hence can provide novel thermoplastic composite materials which particularly is suited for making thin wall articles such as interior fittings for automobiles.

Further, hemp fibers have higher specific heat capacity than synthetic fibers as mentioned before and high thermal bending resistance substantially similar to ligneous celluloses. These characteristics of the hemp fibers can afford to the composites containing the same a good operability and easy handling capability in, for example, stamping process.

Preferred Embodiment For Carrying Out The Invention

In a preferred embodiment of the invention, about 15 to 110 parts by weight of hemp fibers of about .3 to 10mm length are admixed with about 5 to 65 parts by weight of ligneous fillers in the form of particles, dusts or scales of about 0.3 to 10mm size to form an admixture having constituents distributed substantially uniformly, which is added to 100 parts by weight of thermoplastic matrix and dispersed uniformly therethrough to form a novel composite thermoplastic material of the invention. Preferably, the resultant composites are extruded into sheetings for use in stamping process, or pelletized to form pellets for use in injection or transfer molding processes.

Thermoplastic metrices which can be used in the invention include polypropylene, polyethylene, copolymer of ethylene and propylene, acrylonitrile-butadiene-styrene copolymer (ABS), nylon and the like. Among these, polypropylene is most preferred matrix.

Hemp fibers which can be used in the invention include any single fibers selected from ramie, flax, hemp, jute, manila hemp, sisal hemp or the like. In view of stiffness, tenacity or rigidity, and lightweight, manila hemp and jute are particularly preferable reinforcing materials though any limitation is not intended. Lengths of hemp fibers used can be set within wide ranges of from .3mm to 10mm, and preferably about 4mm to 7mm.

Ligneous fillers which can be employed in the invention include those obtained from coniferous or acicular trees, deciduous trees, or crashed hardboard or pulp and other work timber, in the form of particles, scales, chips or dusts, having sizes ranging from .3mm to 10mm, preferably about 2mm to 4mm.

Preferably, hemp fibers are admixed with ligneous fillers in varying proportions including an exemplary ratio of 46 parts by weight of hemp fibers to 65 parts by weight of ligneous fillers. This admixture (110 parts by weight) can be added to 100 parts by weight of polypropylene and kneaded thoroughly to form the composite thermoplastic material of the invention. The resultant composite can be formed into extruded sheetings which are particularly suited for use in stamping operations requiring deep drawing processes to yield excellent products having very high impact strength.

Where particular treatments such as adding lubricant to polypropylene and other thermoplastics, up to 140 parts by weight of hemp single fibers can be added to 100 parts by weight of matrix plastics and yet favorable results can be obtained.

As a comparison, 111 parts by weight of particulate ligneous filler alone are added to 100 parts by weight of polypropylene to form composite thermoplastic material, which is noted as having no uniformity in distribution of fillers and hence poor moldability to inevitably yield many rejects.

Other additives including inorganic fillers, for example, talc can be added in the ratio of about 10 to 40 parts by weight to thermoplastic matrices so as, for example, to improve physical properties such as bending modulus. Appropriate plasticizers and lubricants may be added to the composite material in order to further improve its moldability.

EXAMPLE 1

Into a 500 liter turbomixer, 23kg of manila hemp fibers of about 5mm length and dried to 5 or 6% levels of moisture content are charged. Onto this charge are placed 32.5kg of particulate wood dust screened to particle sizes of 2mm to 3mm dried to about 5% moisture content. The mixer is rotated at 1000rpm for 3 minutes to thoroughly stir the charge. Upon completion of mixing, it is noted that hemp fibers and wood dust are well commingled with each other to form substantially uniform and even admixture.

Then, into this mixer containing the uniform admixture of hemp fibers and wood dust, 50kg of pelletized polypropylene are added portionwise during which the mixer is rotated for 15 minutes at 1500rpm and at temperatures not exceeding 190°C to thoroughly mixing hemp fibers, wood dust and polypropylene.

A resultant mixture is quickly taken out from the mixer and placed in a cooled stirring room wherein the mixture is quenched to form a mass of composite particulate agglomerate, which is then subjected to an ordinary extruder to form sheetings of 2.0mm thickness.

Composite thermoplastic sheetings thus obtained are noted to be particularly suited for use in stamping molding process and to exhibit good handling ability upon heated, good moldability and physical properties and in general well balanced characteristics. In particular, molded articles obtained from novel thermoplastic composites of the invention have high impact strengths at low temperatures and high heat distortion temperatures which have not been expected for prior composites containing ligneous or cellulosic fillers alone, and yet have substantially the same levels of bending modulii and tensile strengths as the prior composites containing ligneous fillers. Novel thermoplastic composites of the invention have little variations in quality throughout their entire mass and can find wider ranges of use than heretofore employed composites having solely ligneous fillers or synthetic fibers, especially for automobile interior fittings such

as instrument panels, door trimming bases with arm rest, center shelves, seat back panels, overhead consoles, heater duct covers and the like.

The same procedures as described above is repeated using jute fibers in place of manila hemp and it is found that substantially the same favorable results are obtained.

EXAMPLE 2

About 55kg of manila hemp fibers dried to 5 or 6% moisture level and having about 5 to 7mm length are placed in the turbomixer. While adding 2.5kg of wood chips of about 3mm particle size and dried to 5 or 6% moisture, the mixer is rotated at 1200rpm for 5 minutes. Upon completion of mixing, wood particles are substantially uniformly distributed through the mass of hemp fibers.

Then, while adding 50kg of polypropylene to the mixer, it is rotated at 1600rpm for 18 minutes at temperatures lower than 193°C.

A resultant mixture thus obtained is quenched to form composite particulate agglomerate and extruded into sheetings in the same manner as in EXAMPLE 1.

Composite thermoplastic sheets in this example contain larger amount of hemp fibers as compared to those of EXAMPLE 1 and exhibit outstanding effects reinforced by fibers. Though they contain only small amount of ligneous fillers, the composite thermoplastic sheets exhibit sufficient rigidity upon heated to permit good

handling ability to be attained in stamping molding process, and to allow the sheets to be worked in double layer due to their tardiness in heat radiation. The sheets of this example are particularly better in deep-drawing moldability than the composites having solely ligneous fillers or synthetic fibers. Though bending modulus is a little lowered, this composite sheeting has highly improved impact strength and moderate heat distortion temperature so that it is suited for use to manufacture deep-drawn articles such as rear quarter panels with arm rests, door trims with arm rests, center shelves, pillar panels in front, center or rear positions.

EXAMPLE 3

7.5kg of ramie fibers having about 3mm length and dried to 5% moisture are first charged in the turbomixer and thereon 32.5kg of wood chips of about 3mm particle size and dried to 5% moisture and 12.5kg of fully dried talc are charged. Then, the mixer is rotated at 800rpm for 3 minutes. At the end of 3 minutes, three constituents are noted to be well mixed up to form uniform admixture.

Into this admixture, 50kg of powdery polypropylene are slowly added and the mixer is rotated at 1500rpm for 15 minutes to stir and knead the content.

A resultant mixture is quenched to form particulate agglomerate, which is then extruded into sheetings as in preceding examples.

This composite sheetings contain very small amount of hemp fibers and have rather lowered impact strength and moldability, but they have good tensile strength, bending modulus and heat distortion temperature. Moreover, this composite sheetings can have sufficient reinforcing effect by ramie fibers and have very little variations in physical properties throughout its entire mass and moderate impact strength thereby to be adapted for use in manufacturing non-deep-drawn articles such as heater ducts, overhead consoles, quarter trims, center shelves, etc.

Industrial Applicability

Advantages attained by the present invention, reside in that, because hemp fibers which are light, rigid and heat-resistant material are employed in admixture with ligneous fillers in the form of particle, dust and scale so as to achieve uniform dispersion of reinforcing materials throughout thermoplastic materials, resultant composite thermoplastics can be free from defects and inadvantages caused by uneven distribution of particulate ligneous fillers of non-uniform shapes and sizes. The composite thermoplastic materials of the invention can be prepared with substantially no variations in physical properties from batch to batch and assure improved and stable physical properties, thermal characteristics and moldability (deep drawability) to be achieved in mass-produced articles with minimum rates of rejects.

In particular, hemp fibers have light weight and yet high rigidity and heat-resistance which would not be expected for other fibers such as synthetic plastic fibers, and exhibit novel reinforcing function in the plastic matrices to thereby impart them improved deep-drawing moldability as well as high impact strength, heat distortion temperature and bending modulus.

Further, hemp fibers have specific heat capacity which is higher than synthetic or artificial fibers and similar to that of ligneous fillers so that composite thermoplastic materials reinforced with hemp fibers can be handled highly stably in the heating process of the stamping molding, and, due to tardy radiation of heat from high specific heat material, novel composite sheetings in double layer or ply can be subjected to stamping moldding process with successful results. The novel composite thermoplastic material of the invention enables a wider range of production control standards including molding conditions to be set because it has excellent and adaptable characteristics and also enables articles with stabilized quality and/or new designs to be produced.

CLAIMS

1. Composite thermoplastic materials comprising 100 parts by weight of thermoplastic matrix, 15 to 110 parts by weight of hemp fibers uniformly dispersed in said matrix, and 5 to 65 parts by weight of ligneous fillers in the form of particle, dust or scale also uniformly dispersed in said matrix.

2. Composite thermoplastic materials as claimed in Claim 1 wherein said thermoplastic matrix is selected from the group consisting of polypropylene, polyethylene, ethylene-propylene copolymer, acrylonitrile-butadiene-styrene copolymer, and nylon.

3. Composite thermoplastic materials as claimed in Claim 2 wherein said thermoplastic matrix is polypropylene.

4. Composite thermoplastic materials as claimed in Claim 1 further comprising from 10 to 40 parts by weight of inorganic fillers.

5. Composite thermoplastic materials as claimed in Claim 4 wherein said inorganic filler is talc.

6. Composite thermoplastic materials as claimed in Claim 1 or 4 further comprising an amount of plasticizer and/or lubricant.

7. Composite thermoplastic materials as claimed in Claim 1, 4 or 6 wherein said hemp fibers are any one selected from the group consisting of ramie, flax, hemp (obtained from Cannabis sativa), jute, manila hemp and sisal hemp.

8. Composite thermoplastic materials as claimed in Claim 1, 4 or 6 wherein said hemp fibers are any two or more selected from the group consisting of ramie, flax, hemp (obtained from Cannabis sativa), jute, manila hemp and sisal hemp.

9. Composite thermoplastic materials as claimed in Claim 1 wherein 46 parts by weight of hemp fibers and 65 parts by weight of particulate, dust or scale ligneous filler are added to 100 parts by weight of polypropylene.

10. Composite thermoplastic materials as claimed in Claim 1 wherein 110 parts by weight of hemp single fibers and 5 parts by weight of ligneous fillers in the form of particle, dust or scale are added to 100 parts by weight of polypropylene.

11. Composite thermoplastic materials as claimed in Claim 1 wherein 15 parts by weight of hemp single fibers, 65 parts by weight of ligneous fillers in the form of particles, dust or scale, and 25 parts by weight of inorganic filler are added to 100 parts by weight of polypropylene.

12. Composite thermoplastic sheetings obtained through extruding the materials as claimed in Claim 1, 9, 10 or 11.

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INTERNATIONAL SEARCH REPORT

International Application No PCT/JP88/00628

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [6]

According to International Patent Classification (IPC) or to both National Classification and IPC

Int.Cl[4] C08L101/00, C08L23/12//(C08L101/00, C08L1:00)

II. FIELDS SEARCHED

Minimum Documentation Searched [7]

Classification System	Classification Symbols
IPC	C08L101/00, C08L23/00

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [8]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [9]

Category *	Citation of Document, [11] with indication, where appropriate, of the relevant passages [12]	Relevant to Claim No. [13]
X	JP, A, 50-60545 (Mitsubishi Chemical Industries Ltd.) 24 May 1975 (24. 05. 75) Page 1, lower right column, line 7 to page 2, upper left column, line 4 (Family: none)	1-12
Y	JP, B2, 54-34020 (Monsanto Co.) 24 October 1979 (24. 10. 79) Column 5, line 1 & DE, B2, 2233011 & FR, B1, 2144807	1-12
Y	JP, B2, 57-54050 (Monsanto Co.) 16 November 1982 (16. 11. 82) Column 7, lines 22 to 24, column 8, lines 15 to 22 & US, A, 3943079 & FR, B1, 2264122	1-12
Y	JP, A, 49-27535 (Nippon Oil Co., Ltd.) 12 March 1974 (12. 03. 74) Page 1, lower left column, lines 5 to 8 (Family: none)	1-12

* Special categories of cited documents: [10]

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
July 25, 1988 (25. 07. 88)	August 8, 1988 (08. 08. 88)
International Searching Authority	**Signature of Authorized Officer**
Japanese Patent Office	